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A MOVING-STRIP FOURIER ANALYZER

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A GRAPHICAL METHOD OF EVALUATING CERTAIN CRYSTALLOGRAPHIC STRUCTURE FACTORS

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Technical Reports 64 to 66
Laboratory for Insulation Research
Massachusetts Institute of Technology

February, 1953

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O.N.R. Contracts N5ori-07801
N5ori-07858

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Abstract: A device embodying Robertson's principle of movable strips is described for performing two-dimensional Fourier summations of the type $\sum_H \sum_K F(hk)[\cos 2\pi(hX + kY)]$ without first rearranging them in the form $\sum_H \sum_K F(hk)[\cos 2\pi hX \cdot \cos 2\pi kY - \sin 2\pi hX \cdot \sin 2\pi kY.]$ The same strips can be used to evaluate sine summations and some three-dimensional summations. Alternatively, the summations can be completed by using masks. The construction of the device, which can accommodate values of $F(hk) = \pm(1 - 99)$, $h = -15$ to 15, $k = 0$ to 15 for X, Y = 1/36 of the repeat distance, is described, and its operation illustrated by an example.

1. Introduction

This analyzer is intended to handle expressions of the form

$$\sum_{-H}^{H} \sum_{0}^{K} F(hk) \begin{matrix}\sin\\ \cos\end{matrix} 2\pi(hX + kY)$$

directly, without first rearranging them in the form

$$\sum_{-H}^{H} \sum_{0}^{K} F(hk)[\cos 2\pi hX \cdot \cos 2\pi kY - \sin 2\pi hX \cdot \sin 2\pi kY].$$

A device embodying this principle was described by Robertson,[1)] and a photograph given showing his moving-strip calculator set up for a two-dimen-

1) J. M. Robertson, Phil. Mag. 21, 176 (1936).

sional summation. Our analyzer differs from Robertson's mainly in that it has been constructed in a form suitable for comparatively inexpensive production, and in that special provision is made for recording and using subtotals. Further, within the limits imposed by inability to accommodate more than $16 \times 31 = 496$ strips at one time in a single outfit, a three-dimensional summation of the form

$$\sum\sum\sum F(hk\ell) \begin{smallmatrix}\sin\\ \cos\end{smallmatrix} 2\pi(hX + kY + \ell Z)$$

can be handled in exactly the same way as a two-dimensional summation.

As a compromise between resolution and compactness, the trigonometric functions are given at intervals of 10° (i.e., 1/36 of the cell edge). In this case, the value at $\pi/2$ is explicit, so that the same strips can be used for both cosine and sine summations. The amplitudes of the strips range from -99 to $+99$. Values of $h = -15$ to $+15$, and of $k = 0$ to 15 are available.

2. The Basic Principle Employed in the Moving-Strip Method

In the discussion which follows, the units of X, Y and Z are 1/36 of the repeat distance. Since all possible values of F(h00) cos $2\pi hX$ are contained on a strip running from 0 to 2π, the first operation is the location of the strips so that the appropriate value is selected for the particular point (X) at which the summation is being evaluated. As an example, consider the strip of amplitude 93:

	93	93	92	87	81	71	60	46	32	16	0	$\overline{16}$	$\overline{32}$	$\overline{46}$	... etc.,
where hX =		0	1	2	3	4	5	6	7	8	9	10	11	12	... etc. (I)

If h = 1, then, as X increases, successive values of 93 cos $2\pi hX$ will be those printed on the strip; if h = 2, then successive values of cos $2\pi hX$ will be for

X = 0 . 1 . 2 . 3 . 4 . 5 . 6 ... etc., (II)

and when h = 3, for

X = 0 . . 1 . . 2 . . 3 . . 4 ... etc. (III)

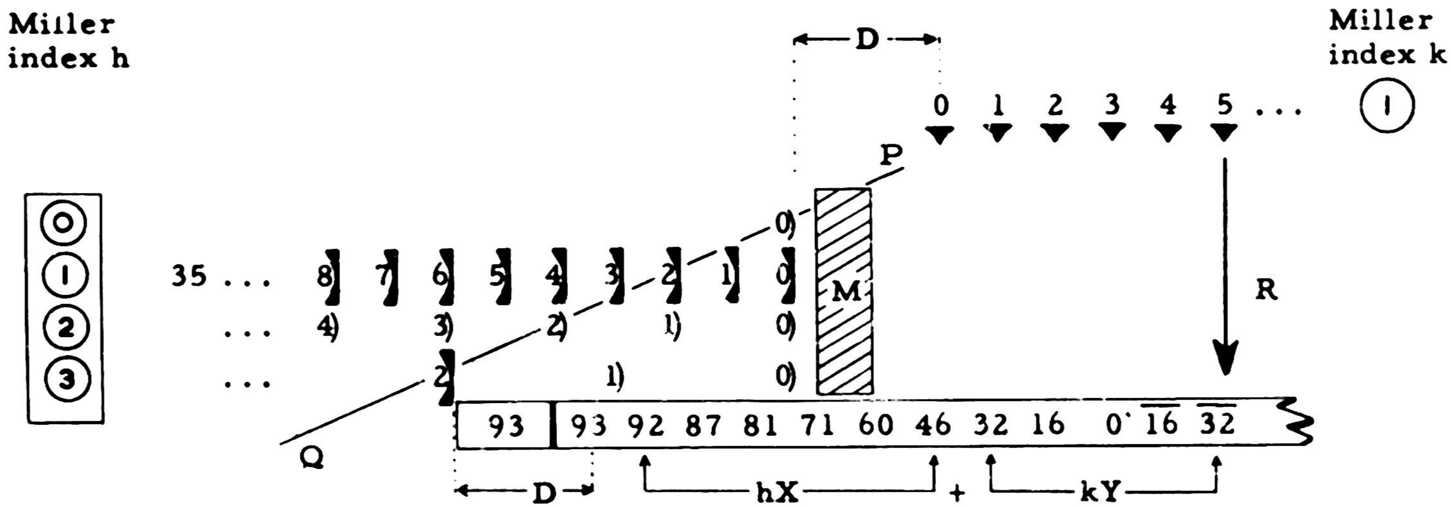


Fig. 1. Basic principle for the summation of a one-dimensional series of the form $\sum_{0}^{H} F(hk) \cos 2\pi(hX + kY)$ when k and X are kept constant. Here k = 1, X = 2.

Hence, if an arbitrary origin (i.e., M in Fig. 1) is chosen, and a set of scales like (I), (II), (III) are marked off from this origin, reading to the left, the value of F cos $2\pi hX$ can be obtained as follows: Suppose, as in Fig. 1, that amplitude F(31) = 93. The strip "93" is chosen and inserted in the slot corresponding to h = 3. Suppose that the electron density is to be evaluated at the point (X, Y) = (2, 5). Since X = 2, the left-hand end of the strip is set against the number "2" on the location scale, and the value of F(31) cos $2\pi(hX + kY)$ will appear under the number "5" (since Y = 5) on the right-hand side of the origin M. Thus the value of 93 $\cos(2\pi/36)(3 \times 2 + 1 \times 5) = \overline{32}$. It will be seen that if the one-dimensional summation having constant k, such as

$$\sum_{0}^{H} F(h1) \cos 2\pi(h \cdot X + 1 \cdot Y) \qquad \text{(IV)}$$

is to be evaluated at the point (X, Y) = (2, 5), the ends of the strips of amplitudes $F(h\ell)$ will lie along the line PQ, thus automatically displacing each strip a distance hX (i.e., 2h) to the left of the origin. The value of kY at any point

(X, Y) is constant for this group of terms of constant k, and hence the value of one-dimensional summations like (IV) at the point (2, 5) is obtained by summing the numbers in the vertical column R. Without altering the setting of the strips (i.e., keeping the value of X constant) this one-dimensional summation having constant k can be obtained for all values of Y (i.e., at all the points (2, Y)).

A two-dimensional cosine series can therefore be handled as a set of one-dimensional summations, each of constant k; and if these are arranged on a set of boards each bearing a k location scale on the right-hand side (which scale will be the mirror image of the corresponding h location scale on the left-hand side) then the strips having amplitudes corresponding to all the observed values of F(hk0) can be inserted on the boards at the beginning of the operation and used throughout the preparation of the projection.

A calculator embodying this principle must therefore meet the following requirements: (1) As can be seen from Fig. 1, a separate board will be needed for each value of the Miller index k, bearing the appropriate k-location scale along the top edge, while the set of h-location scales, which will be called the Location Sheet, will be the same for all boards; (2) Since values of the cosine function must appear in all columns like R in Fig. 1, even when the end of the strip is located at the left-hand side of Fig. 1, a strip must carry two complete cycles of the cosine function; (3) Some kind of guide must be provided to keep the strips in place; (4) If the cosine strips are to be used for sine summations, it must be possible to move the location sheet; thus two complete cycles of these h scales will be needed, as for the strips; (5) Provision must be made for recording the totals of the one-dimensional summations, preferably in a form which will be convenient for future use.

3. Details of Construction

In order to obtain clear and easily legible numbers, the original strips

and location scales were prepared on an I.B.M. "Executive" electrical typewriter; the resulting appearance of the printed version can be seen in Fig. 1. Available facilities fixed the optimum size of the printed cards at 17 in. x 11 in., which is therefore the size of the location sheet, while the cosine strips are 17 in. long. Such a strip, even when cut from heavy card, will be too flimsy if it is less than 1/4 in. wide and it was not possible to support for easy movement more than 3 such strips per inch. The working height of the location sheet being approximately 10 in., the maximum number of strips was thereby fixed at 31, i.e., h = –15 to +15. This limitation is unfortunate, since in fact 18 orders should be available if the cell edge is divided into 36 parts.

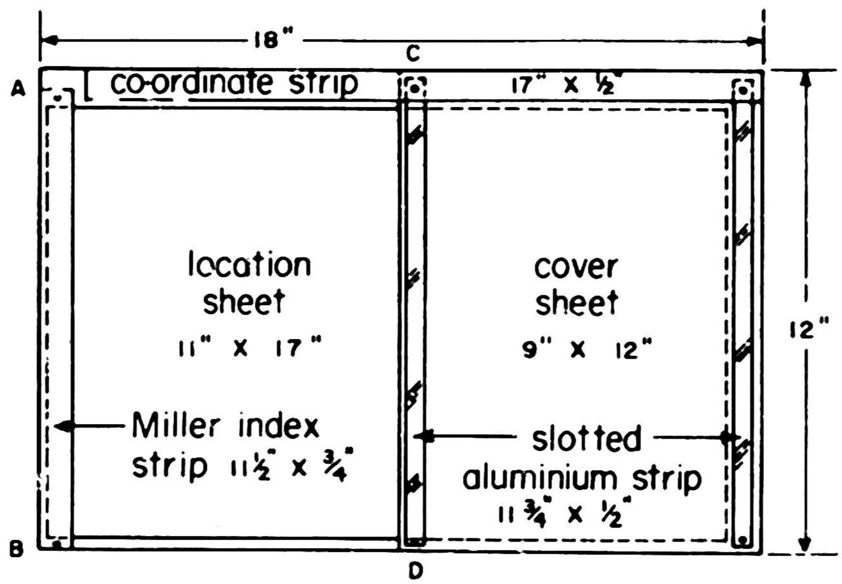


Fig. 2. Layout of the boards used for summations.

To meet requirements (1) to (5), the design shown in Fig. 2 was adopted. The backing board is a 12 in. x 18 in. sheet of white-coated, pulp board to which the other components are affixed with staples. The co-ordinate strip and the location sheet are printed in red on high-grade rag card (175 lbs). The Miller index strip and the cosine strips are printed in black on a heavier card (200 lbs.) of the same type. The cover sheet, which prevents location-sheet numbers ap-

pearing behind the cosine strips, is a 24-lb. white paper. Requirement (3) is met by making the cosine strips slide in slots stamped in 2S0 aluminum sheet, 1/2 in. wide and 0.012 in. thick, by means of a rotary stamp (Fig. 3). Requirement (4) is met by sliding the location sheet under the Miller index strip and under the slotted strip stapled to the backing board at A and B, and at C and D, respectively. Requirement (5) is met by providing a spare slot for a 1/4 in. strip just below the K scale.

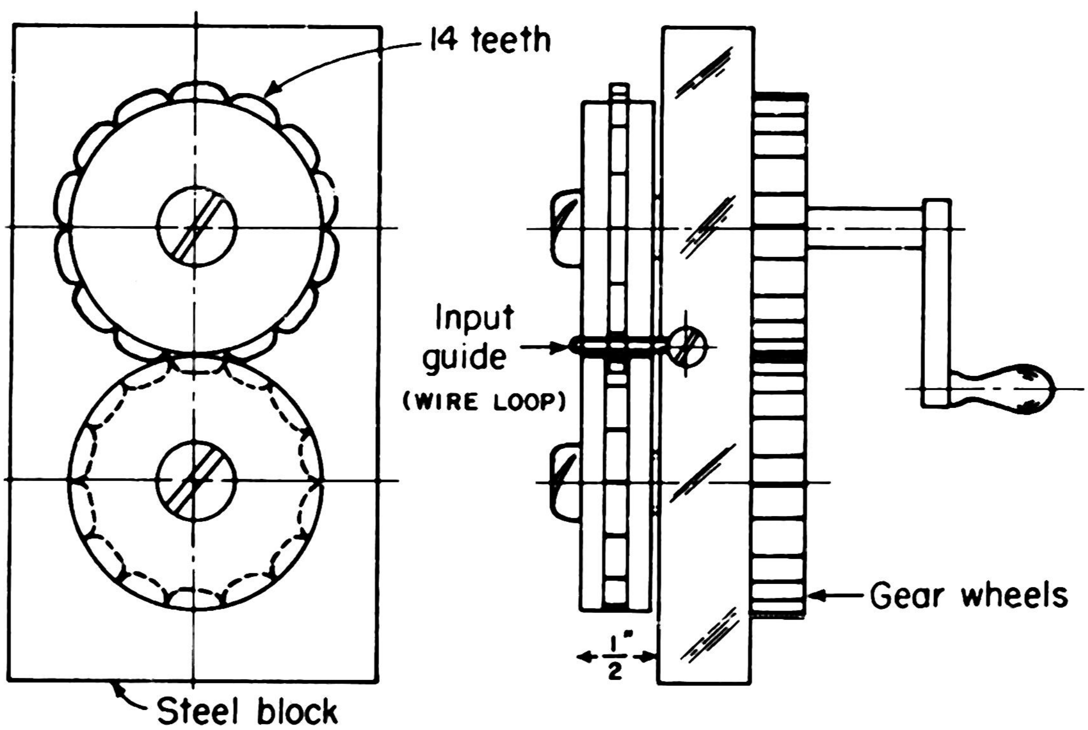


Fig. 3. Rotary stamp used for slotting the aluminum strip.

There are 16 boards (k = 0 to 15) of the type described, 5 sets of strips having positive and negative amplitudes 1 to 33 and 3 sets of strips having + and – amplitudes 34 to 99, a total of 726 strips, which are already cut. One complete set of strips of positive and negative amplitudes 1 to 99 are laid out in flat files, similar to that shown in Fig. 2 of Robertson's paper.[2)]

2) J.M. Robertson, J. Sci. Instr. 25, 28 (1948).

4. Evaluation of a Two-Dimensional Projection

As an example of the method, the electron density in the two-dimensional [010] projection of a 1, 4-dimethoxybenzene molecule is computed from data published by Goodwin, Przybylska and Robertson.[3] The space group is Pbca, so that (h0ℓ) reflections occur only when ℓ = 2n. The observed F(h0ℓ) data contained in Table 5 of their paper have been used to derive the values shown in Table 1, which are the amplitudes actually used in the summation $\rho(X, 0, Z) = \sum_{0}^{H}\sum_{0}^{L} F(h0\ell) \cos 2\pi(hX + \ell Z) + \sum_{-H}^{0}\sum_{0}^{L} F(\bar{h}0\ell) \cos 2\pi(-hX + \ell Z)$. In the ensuing discussion and in Figs. 4 and 6 only the first term of this expression has been used for illustrative purposes, but Fig. 7b and hence Table 2 were obtained from the complete expression.

This example was chosen because, in the unit cell, a = 7.29Å, c = 16.55Å, so that reflections were observed for which $\ell \leq 20$; hence it was possible to see whether, for practical purposes, values of $\ell > 15$ could be neglected, as must be done with this moving-strip calculator.

The origin was taken at a center of symmetry, and the projection plotted for X =($2\pi/36$)(0 to 9), Z = ($2\pi/36$)(0 to 18). Since $Z_{max} > X_{max}$, the one-dimensional summations of constant $\underline{\ell}$ are evaluated rather than those of constant $\underline{h}$. One board is therefore required for each of the 8 values of ℓ = 0, 2, ...14; strips of amplitudes given in the appropriate column of Table 1 are inserted in the correct $\underline{h}$ slots on each board, as can be seen in Fig. 4 on the board for which ℓ = 14. To calculate the electron density at a row of points, say (3, 0, Z), the left-hand end of each strip is placed against the marker for "3" on its $\underline{h}$-location scale, and the summations performed for each column shown to be necessary by the markers on the ℓ scale. Since cos (A + π) = —cos A, it is never necessary to compute more than 18 out of the possible 36 subtotals on any board, and often fewer are needed; for instance, in the example chosen, in which ℓ = 2n, it can be seen from the scales for all the 8 boards visible in Fig. 4 that only

3) T.H.Goodwin, M.Przybylska, and J.M.Robertson, Acta Cryst. $\underline{3}$, 279 (1950).

Table 1. Values[4] of $F(h0\ell)$ used to obtain a projection of a 1,4-dimethoxybenzene molecule on (010). a = 7.29, c = 16.55Å. The number of electrons / unit cell = 296 is taken as F(000), the values of F(h00) and $F(00\ell)$ have been multiplied by 2, and those of $F(h0\ell)$ by 4 (on account of the multiplicity of these reflections). All values have then been divided by the area of the (010) projection, so that the computed values of $\rho(X, 0, Z)$ will be on the scale 100 = 1 electron/$Å^2$. Values of $F(h0\ell)$ with $\ell > 14$ have been ignored.

4) Based on data published by T.H. Goodwin, M. Przybylska, and J.M. Robertson, Acta Cryst. 3, (1950) 279.

h \ ℓ	0	2	4	6	8	10	12	14	16	18	20
0	245	56	$\overline{41}$	55	$\overline{40}$	$\overline{36}$	44	19	$\overline{2}$	3	$\overline{4}$
1		12	$\overline{76}$	22	45	18	41	23	3	8	
2	160	25	$\overline{88}$	43	$\overline{50}$	$\overline{50}$	60	33	$\overline{12}$		$\overline{5}$
3		$\overline{13}$	$\overline{66}$	12	23	25	60	28	3	11	
4	14	$\overline{46}$	$\overline{80}$	$\overline{7}$	$\overline{8}$	$\overline{23}$	26	20	$\overline{21}$	$\overline{11}$	
5		$\overline{7}$	$\overline{7}$	5	3		13				
6	$\overline{2}$	$\overline{25}$	$\overline{38}$	$\overline{15}$		$\overline{5}$	12	10	$\overline{12}$		
7		$\overline{5}$	13	7	$\overline{12}$	$\overline{8}$	$\overline{15}$	$\overline{13}$			
8	9	5	$\overline{8}$	$\overline{7}$	7						
9			7								

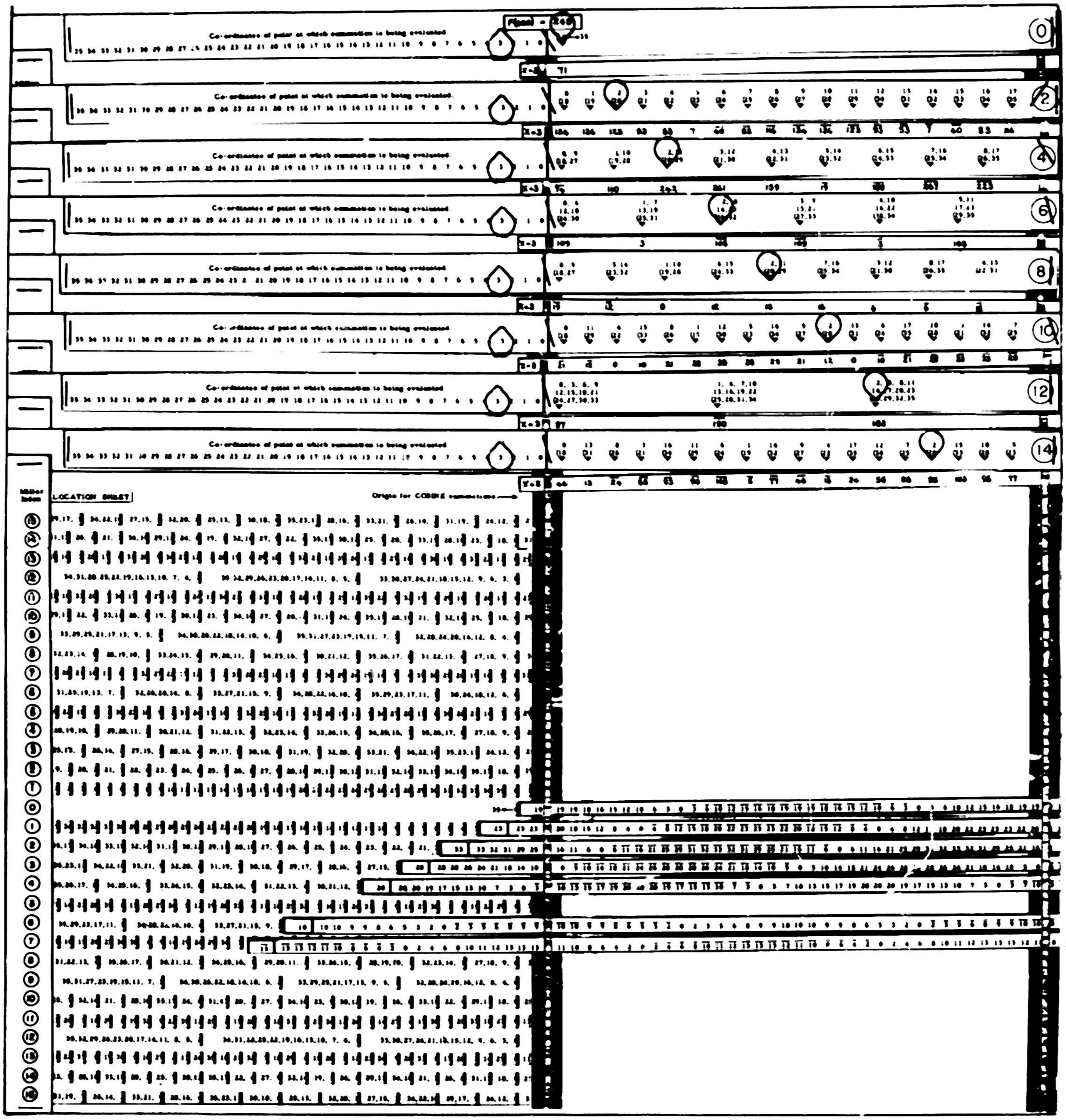


Fig. 4. (a) Evaluation of the one-dimensional series $\sum_{0}^{M}$ F(h, 14) cos 2π(h, 3 + 14.Z) at the points (3, 0, Z). On the board ℓ = 14, strips of amplitudes F(h, 0, 14) obtained from Table I, are placed in the h slots. Since X = 3, the left-hand end of each strip is set against the number "3" on the Location Sheet, and the subtotals for each value of Z written on the subtotal strip in the top slot. Such a strip for X = 3 is prepared for each value of ℓ, i.e., for each board. (b) Completion of the ρ (3, 0, Z) summation. If the boards are arranged in a staggered pile as shown, the final summation at any point (3, 0, Z) can be picked out. In the figure the indicators are set to pick out the summation at (3, 0, 2).

$$1 + 9 + 9 + 3 + 9 + 9 + 3 + 9 = \underline{52 \text{ subtotals}}$$

have to be computed for each value of $X \neq 0$. The strips must be reset each time X is changed, i.e., for every line of constant X in the projection.

5. Methods of Completing the Two-Dimensional Summation

A. Evaluation of $\rho(X, 0, Z)$ at each point in turn. One especially useful feature of this method is that the electron density at any selected point (X, 0, Z) can be obtained independently by setting all the strips for the value "X" on the h scales, and adding together the "Z" column from each of the 8 boards without prior evaluation of subtotals. If the electron density is required at a small number of arbitrarily chosen points in the projection, this method is very fast. If, however, the values of $\rho(X, 0, Z)$ are required for the complete projection, it is better to make use of subtotals for two reasons:

1. For every value of ℓ which is a factor of 36, or contains a factor of 36, the same column of numbers will be used for two or more values of Z. For example, when $\ell = 12$, the three columns indicated in Fig. 4 are each used for 12 values of Z. It is a considerable waste of time to sum these columns again for each of these 12 values, especially when there are a large number of strips on the board.

2. An appreciable time is inevitably consumed in the manipulation of strips and boards, and this is reduced to a minimum by using subtotals.

B. Use of subtotal sheets. In Fig. 4 it can be seen that when the subtotals have been computed for a given value of X, the final summation at any point (X, 0, Z) will be obtained by summing the appropriate subtotals, which can be picked out on the subtotal strips. In Fig. 4 the subtotals required for $Z = 2$ are shown by ring indicators. It is quite possible to complete a summation in this way, but the staggered pile of boards becomes unwieldy when all 16 boards are in use; hence a subtotal sheet (reproduced in Fig. 5) containing these

SUBTOTAL SHEET

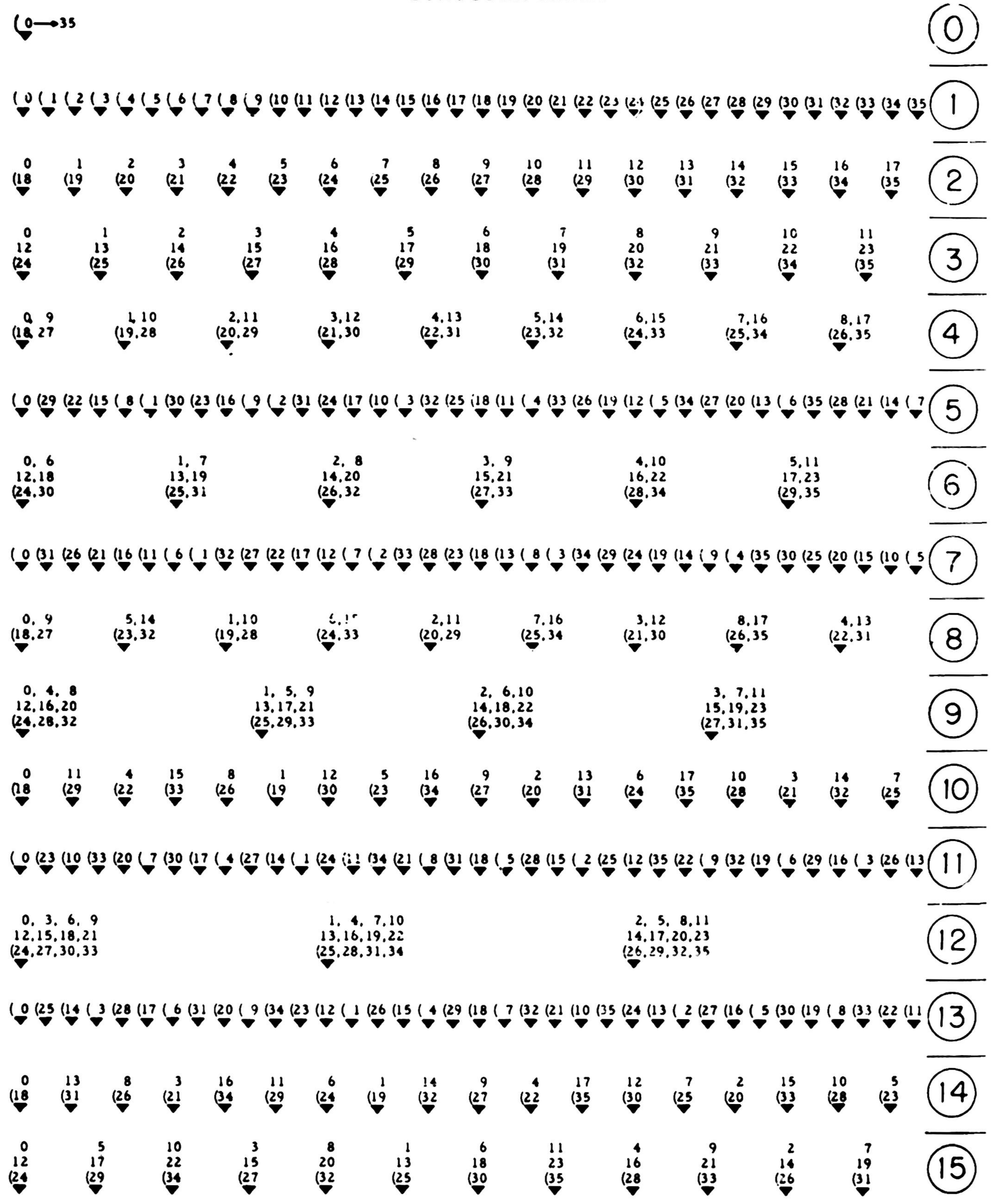


Fig. 5. Location scales for 0, 1, 2. . . . 15. When all 16 boards are in use, the staggered pile (Fig. 2) becomes unwieldy. If a sheet of tracing paper is superposed on Fig. 5., the subtotals for a particular value of X can be entered on it, and the final totals prepared from this instead.

scales is supplied with the device: the subtotals can be written on a sheet of tracing paper superposed on these scales, and the final totals prepared from this instead; the tracing can then be kept for reference.

Keeping such a record of the subtotals for each value of X has an advantage if, in successive Fourier summations, the sign of an $F(h0\ell)$ term has to be changed, and this is done by adding a term of opposite sign having twice the amplitude of $F(h0\ell)$. The appropriate cosine strip can be inserted in the second spare slot, directly beneath the subtotal strip, with its left-hand end located at the required value of X in the usual way, and then the corrected subtotals are obtained merely by adding the two strips together.

This designation of subtotals by means of ring indicators, as in Fig. 4, is equivalent to the use of masks for the selection of terms.[2, 5] Although they could easily be made, no masks are provided with this device because it is felt that while they would certainly reduce the probability of error, masks are not absolutely necessary as the value of Z is explicitly stated above each subtotal.

C. <u>Subtotal strips treated like cosine strips</u>. On the board for which $\ell = 0$, there will be space for strips of amplitudes $F(h00)$ and $F(\bar{h}00)$. Since, in general, $F(h00) \equiv F(\bar{h}00)$, only one of these groups will be needed, and either set of slots may be used.

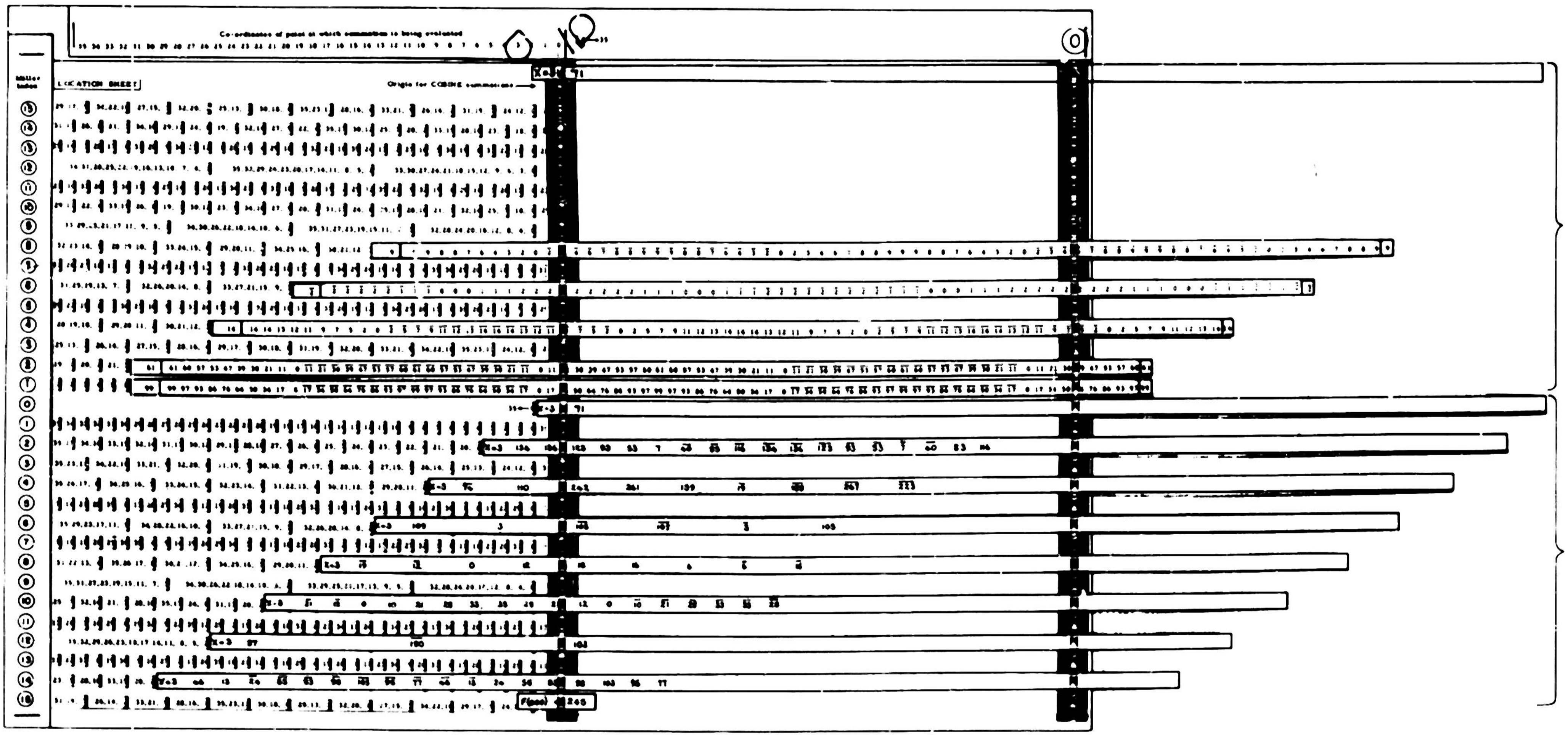
If, therefore, the $F(\bar{h}00)$ slots are used, as in Fig. 6(a), then the $F(h00)$ slots will be available for use with the subtotal strips. All the subtotal strips visible at the top of Fig. 4 are placed on the board for which $\ell = 0$, the strip from the board $\ell = \underline{n}$ being placed in the slot for which $h = \underline{n}$. The summation at all the points (3, 0, Z) in turn is obtained by setting the strips against the number "Z" on the location sheet, and summing the numbers in the first column to the right of the origin (M in Fig. 1). The strips in Fig. 6 are set to give the value of $\rho(3, 0, 2) = 807$, which, as stated in Table 1, is equivalent to $8.07 e/\mathring{A}^2$.

5) A. L. Patterson and G. Tunell, Amer. Min. <u>27</u>, 655 (1942).

Fig. 6. Evaluation of the summation $\rho(X, 0, Z) = \sum_0^H \sum_0^L F(h0\ell) \cos 2\pi(hX + \ell Z)$ using the board for which $\ell = 0$.

a) Evaluation of the one-dimensional series $\sum_0^H F(h00) \cos 2\pi hX$, where $X = 3$. For this summation it is possible to use the $\bar{h}$ slots instead of the h slots, since $\cos 2\pi(\bar{h}X) = \cos 2\pi(hX)$. The value of $F(200) = 160$ is obtained by using strips of amplitudes 99 and 61 together (possible only when there is an empty slot next to the F(200) slot). The value of this one-dimensional series for $X = 3$ (i.e., 71) is written on the subtotal strip in the top slot.

b) Evaluation of $\rho(X, 0, Z)$ for $X = 3$, $Z = 2$, using the subtotal strips for $X = 3$. The subtotal strip for $X = 3$, $Z = 0 - 35$ is taken from each board for which $\ell = n$ (see Fig. 2) and inserted in the Miller index slot $\underline{n}$ as shown. The end of each strip is then set against "2" on the location sheet (since $Z = 2$), and the sum of the numbers in the first column to the right of the origin (M in Fig. 1) is the value of $\rho(3, 0, 2)$.



Because a certain amount of time is inevitably consumed in manipulating the boards and strips, it is difficult to estimate the time taken for a summation. The relative speeds of the alternatives already described depend partly on the number of strips in use on each board, and partly on the shape of the section of the total projection which has to be computed. For example, when subtotals are used, if all the possible subtotals for a fixed value of X are computed for each board in turn (the fastest method of subtotalling), this will in general have to be done regardless of whether Z runs from 0 to 35, or only 1/2 or 1/4 this range. Also, it has to be done again for each value of X, so that the subtotalling method is much faster (indeed, nearly twice as fast if the boards are well-filled) when the projection has to be evaluated at, say, Z = 0—35, X = 0—8, than when it must be done for Z = 0—17, X = 0—17, although the summation is required at the same number of points.

6. Accuracy and Applicability of the Method

The completed projection obtained from this summation is shown in Fig. 7, together with the projection published by Goodwin et al.[3)] for comparison. The general shape of the molecule is easily recognizable, and the co-ordinates of the atomic centers, taken merely as the estimated positions of the highest points of the peaks on a contour map on the scale 1 inch = 1Å, are given, together with the published values, in Table 2. In view of the fact that some of the high-order terms neglected were quite large (up to 13 percent of F_{max}), and that no attempt was made to estimate the true centroid of a peak, the excellent agreement obtained in some cases is probably fortuitous.

This raises the following question: suppose atomic co-ordinates estimated from a Fourier projection on a rather coarse mesh (as has been done here) without elaborate attempts to obtain high accuracy, are used to compute structure factors, and that these give a value of $R \simeq 30\%$; will this be a satis-

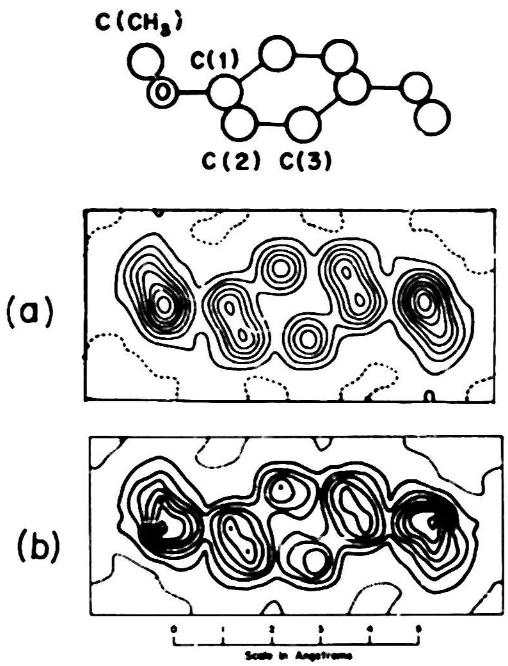


Fig. 7. [010] projection of 1,4-dimethoxybenzene molecule; (a) from data of Goodwin et al.[3]; (b) as computed from data in Table 1, where $\ell \leqslant 14$.

factory basis for the application of a least-squares refinement process? Work now in progress on the least-squares method suggests that this will be true in at least a certain number of cases. If so, a Fourier summation device of the type described, with values obtainable at 10° intervals, would have a wider application than might otherwise be expected. In any case it will be adequate for work on crystals in which reflections with h, k, $\ell > 15$ are not observed.

7. Evaluation of Series Other than the Two-Dimensional Cosine Series Involving $\cos 2\pi(hX + kY)$.

Two important extensions of the method are indicated in the caption to Fig. 8. In addition to these, summations of the form $\cos 2\pi(hX - kY)$ can be obtained by placing the F(hk) strips in the $\bar{h}$ slots, since $\cos 2\pi(-hX + kY) = \cos 2\pi(hX - kY)$.

Another possibility is the inclusion of phase angles. Robertson[1] suggested that for noncentrosymmetric projections, an additional displacement of the strip corresponding to the phase angle could be used, with the limitation that it can be varied only in discrete amounts (multiples of 10° in this analyzer).

It is also possible to carry out the Beevers-Lipson type of summation[6] involving expressions like $F(hk\ell) \frac{\sin}{\cos} 2\pi hX \cdot \frac{\sin}{\cos} 2\pi kY \cdot \frac{\sin}{\cos} 2\pi \ell Z$ by evaluating $F(hk\ell) \frac{\sin}{\cos} 2\pi hX$, and using this as a new amplitude $F'(hk\ell)$, and so on. In this case the one-dimensional summations required can be obtained by locating the strips as usual and evaluating $\cos 2\pi(hX + kY)$, where $Y = 0$. Alternatively, if

6) C. A. Beevers and H. Lipson, Proc. Phys. Soc. 48, 772 (1936).

Fig. 8. Extension to series other than the two-dimensional cosine series $\cos 2\pi(hX + kY)$.

a) SINE summations: Because $\sin 2\pi(hX + kY) = \cos\left[2\pi(hX + kY) + 3\pi/2\right]$, when the Location Sheet is moved $3\pi/2$ to the left, as illustrated, and the strips located in the same way as for cosine summations, sine summations will be obtained. The strips as set here would evaluate $\sum_{0}^{H} F(hk) \sin 2\pi(hX + 2Y)$ at the points $(1, Y)$.

b) THREE-DIMENSIONAL summations: If the Location Sheet is moved a distance ℓZ, i.e., $3 \times 8 = 24$ in the example given below (instead of $3\pi/2$, as for sine summations) then strips located in the ordinary way will permit the summation of series such as $\sum_{-H}^{H} F(h43) \cos 2\pi(h7 + 4Y + 3 \times 8)$ at the series of points $(7, Y, 8)$ on the board for which $k = 4$. The ℓZ shift is obtained by putting the origin at the required value of Z on the $-h$ strip.

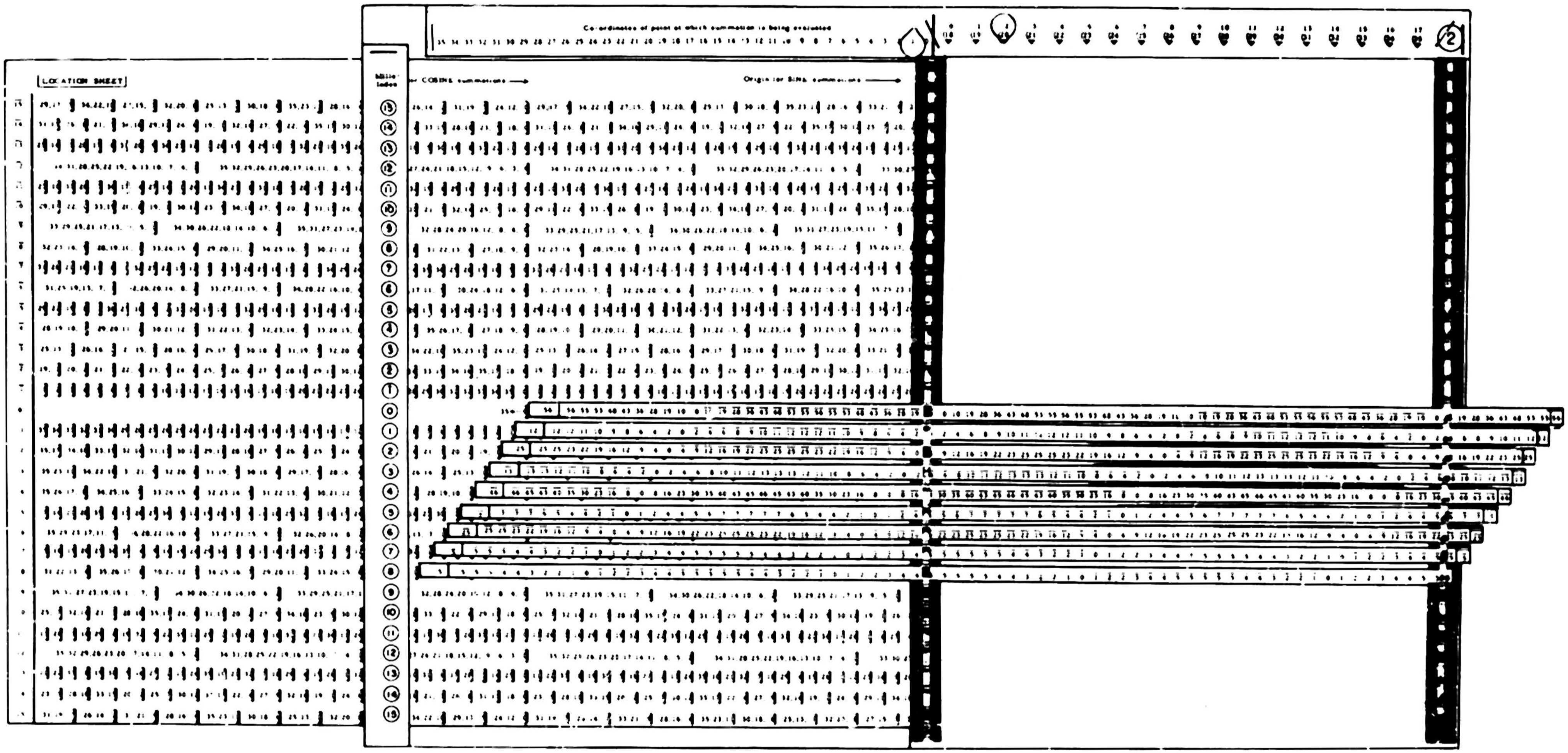


Table 2. Co-ordinates x and z of atoms in 1,4-dimethoxybenzene molecule.

Atom	G.P.& R. x	G–W x'	Δx = x–x'	G.P.& R. z	G–W z'	Δz = z–z'
C(1)	0.0055	0.008	–0.002	0.0777	0.082	–0.004
C(2)	0.0927	0.074	+0.016	0.0612	0.061	0
C(3)	0.0977	0.108	–0.011	–0.0156	–0.021	+0.005
0	0.0104	0.010	0	0.1541	0.164	–0.010
$C(CH_3)$	–0.0763	–0.069	+0.007	0.1730	0.166	+0.007

all the strips were set for X = 0, they would present the same appearance as Robertson's sorting board,[2)] except that values of the cosine function are listed for the whole cycle 0 to 2π instead of only 0 to $\pi/2$, so that the sign of each term is explicit and does not have to be supplied by auxiliary markers. It would therefore be possible to pick out the summation by placing the strips in the subtotal slots, and proceeding as in section 5(b), or by making masks. However, since devices are obtainable which have been specifically designed for computing Fourier summations arranged as sine and cosine products, this represents a need which has already been satisfactorily met, and therefore no masks have been provided.

Acknowledgements

The production of this device in quantity has been possible only through the co-operation of several people. I am especially grateful to Professor A. L. Patterson for both moral and financial support; to Mrs. Hariot B. Armstrong, who typed the originals for reproduction; to Mr. A. J. Ginty for help in the selection of suitable papers; to Mr. D. G. Webster of the M.I.T. photo service for valuable advice, and to Mr. A. Pellecchia, also of the M.I.T. photo service, without whose care for the accurate printing and trimming of the strips the project would have been impossible; to Mr. P. Kelleher for making the rotary

stamp shown in Fig. 3; I also wish to thank Professor A. von Hippel for his interest in the work, Dr. S. C. Abrahams for helpful discussions, and Mr. S. M. Kingsbury for advice on the technical aspects of the project; and Mrs. C. Grenville-Wells and Miss J. Lonsdale for assistance in the tedious job of assembling the completed outfits.